1. Introduction

In the field of fluid analysis, many studies have used particle methods such as SPH (Smoothed Particle Hydrodynamics)\(^1\) and MPS (Moving Particle Semi-implicit)\(^2\). In particular, particle methods are often used in situations where the topology of the fluid changes dynamically. Such situations are difficult to be analyzed with FVM (Finite Volume Method) or FEM (Finite Element Method) because neither method can treat dynamic topological change. Then, some applications use particle methods such as simulations of waterfalls, avalanches, and so on.

We have also tried a simulation using a particle method whereby we injected viscous fluid from a catheter into a water tank in order to investigate the droplet formation process. Previous studies used an interfacial tension model that could consider the influence of the other phase in two-phase flow. However, the environment of the physical experiment differed from that of the simulation. Therefore, in this study we performed a viscous fluid injection simulation using the same environment as in the physical experiment. On the basis of the results, we have validated the proposed method by comparing the droplet size and the formation cycle between the physical experiment and the simulation.

2. Related works

A number of related works with particle methods bear mentioning. Yuhashi et al.\(^7\) used MPS method to investigate the resistance to stirring in a camshaft. Duan et al.\(^8\) simulated the diffusion of highly viscous fluid with MPS method to model crust formation. Saso et
al.\textsuperscript{9)} analyzed a molten site with a MPS-based welding simulation by considering not only surface tension but also Marangoni force. Shino et al.\textsuperscript{10)} performed a press molding analysis of CFRT (Carbon Fiber Reinforced Thermo Plastics) with MPS method. Furthermore, particle methods are applied not only to industrial fields but also to medical fields. Gambaruto used MPS method to analyze blood flow with red blood cells in small veins\textsuperscript{11)}, and Kikuchi et al.\textsuperscript{12)} used MPS to simulate the human swallowing function. In addition, the accuracy of MPS method has increased thanks to certain studies. Tanaka et al. improved the accuracy of pressure calculation by adding the velocity divergence term to the source term of Poisson equation in MPS method\textsuperscript{13)}. Iribe et al. improved the accuracy of gradient calculation\textsuperscript{14)}, and Tamai et al. developed a highly precise spatially discrete scheme of MPS method using the least squares method\textsuperscript{15)}. Moreover, Matsunaga et al.\textsuperscript{16)} improved the precision of the wall boundary for the highly precise spatially discrete scheme developed by Tamai et al.

However, it is difficult to apply these methods to a case where a droplet is formed in a different type of fluid, because the above methods were developed for specific cases. Therefore, we have developed a method that can be applied to the viscous fluid injection simulation in liquid-liquid two-phase flow.

3. Analytical method

3.1 Governing equations

In this study, the viscous fluid injected through a circular tube imitating a catheter and the water that fills the tank are treated as incompressible fluids. The governing equations for incompressible fluid are the Navier-Stokes equation and the equation of continuity as the mass conservation law, which are defined in the following.

\[
\frac{Du}{Dt} = -\frac{1}{\rho} \nabla P + \nu \nabla^2 u + g + \frac{1}{\rho} F_{\text{inter}} \tag{1}
\]

\[
\frac{D\rho}{Dt} = 0 \tag{2}
\]

where \(\rho\) is the density of the fluid, \(t\) is time, \(u\) is the velocity vector, \(P\) is pressure, \(\nu\) is the kinematic viscosity coefficient, \(g\) is the gravitational acceleration vector, and \(F_{\text{inter}}\) is the interfacial tension. On the right of Eq. (1), the first four terms are pressure, viscosity, gravitational acceleration, and interfacial tension terms, respectively.

3.2 MPS method

MPS method treats the particle interaction by discretizing the partial differential equation, which uses a weight function to calculate the weighted average of interparticle distance. In this study, we use the weight function developed by Tamai et al.\textsuperscript{15)}, which is defined as follows.

\[
\omega(r_{ij}) = \begin{cases} 
\left( \frac{r_{ij}}{r_e} - 1 \right)^2 & (r_{ij} \leq r_e) \\
0 & (r_e < r_{ij})
\end{cases} \tag{3}
\]

where \(r_{ij}\) is the distance between particle \(i\) and \(j\), and \(r_e\) is the radius of a particle’s influence. In this study, \(r_e\) is set as \(2.1\) times the initial interparticle distance in the gradient and the divergence models, and as \(3.1\) times in the Laplacian and the interfacial tension models because it is generally set as \(2\) to \(4\) times the initial interparticle distance. MPS method requires the calculation of the density of a particle to keep it constant. The density of a particle is calculated with Eq. (4), which uses Eq. (3).

\[
n_i = \sum_{j \neq i} \omega(r_{ij}) \tag{4}
\]

where \(n_i\) is the particle number density of particle \(i\).

In the particle interaction models, the gradient, the divergence, and the Laplacian models of particle \(i\) are defined in the following differential operators.

\[
\langle \nabla \phi \rangle_i = \frac{d}{n^0} \sum_{j \neq i} \frac{\phi_j - \phi_i}{r_{ij}^2} (r_j - r_i) \omega(r_{ij}) \tag{5}
\]

\[
\langle \nabla \cdot \phi \rangle_i = \frac{d}{n^0} \sum_{j \neq i} (\phi_j - \phi_i) \cdot (r_j - r_i) r_{ij} \omega(r_{ij}) \tag{6}
\]

\[
\langle \nabla^2 \phi \rangle_i = \frac{2d}{\lambda n^0} \sum_{j \neq i} (\phi_j - \phi_i) \omega(r_{ij}) \tag{7}
\]

where \(\phi\) is an arbitrary scalar value, \(d\) is the dimension number, \(n^0\) is the initial particle number density, \(r\) is the particle coordinate vector, \(\phi\) is an arbitrary vector, and \(\lambda\) is the coefficient of the Laplacian model, which is defined in the following.

\[
\lambda = \frac{\sum_{j \neq i} r_{ij}^2 \omega(r_{ij})}{\sum_{j \neq i} \omega(r_{ij})} \tag{8}
\]

3.3 High order MPS method

MPS method described in the Section 2.2 is the original model developed by Koshizuka et al.\textsuperscript{2)}, which assumes that particles are regularly arranged. The calculation becomes unstable when the particle arrangement is imbalanced. Then, in this study, some high-order MPS methods are adopted to stabilize the calculation even in an imbalance arrangement of particles.

Firstly, in the Poisson equation for pressure
calculation, we adopt a model with the velocity divergence term developed by Tanaka et al.\textsuperscript{13}) whose source term is as follows.

\begin{equation}
\langle \nabla^2 p \rangle_j^{k+1} = \frac{\rho}{\Delta t} \nabla \cdot \mathbf{u}_i^* + \alpha \frac{\rho}{\Delta t^2} \left( n_j^k - n_i^k \right)
\end{equation}

where $\Delta t$ is the time step size, $\mathbf{u}_i^*$ is the provisional velocity vector, $\alpha$ is the relaxation coefficient, and $n_j^k$ is the particle number density at the time step $k$. $\alpha$ is a coefficient that is set depending on the problem, and in this study, it is set as 0.02 according to the pre-calculation result.

Secondly, we adopt the high order gradient model developed by Iribe et al.\textsuperscript{14}). Furthermore, to prevent the excessive approach of particles, we use a model developed by Monaghan\textsuperscript{17}) that considers the artificial repulsive force that is added to the gradient model developed by Iribe et al.\textsuperscript{14}). The pressure gradient is defined in the following.

\begin{equation}
\langle \nabla P \rangle_j = \left[ \frac{1}{n_i^0} \sum_{j \neq i} \frac{(r_j - r_i)}{r_{ij}} \otimes \frac{(r_j - r_i)}{r_{ij}} \omega(r_{ij}) \right]^{-1}
\end{equation}

where $\otimes$ is the tensor product, and $\hat{P}_j$ is the minimum pressure in the radius of influence so that $\hat{P}_j$ is always lower than $P_j$. Since $P_j - \hat{P}_j$ is always positive, the repulsive force is generated between particles $i$ and $j$. Therefore, it is possible to prevent the excessive approach of particles due to the attraction. Eq. (10) enables the calculation stable by replacing $d$ with the inverse matrix in Eq. (5) even when particles are not regularly arranged. The inverse matrix becomes a unit matrix when particles are regularly arranged, in which Eq. (10) is equivalent to Eq. (5).

### 3.4 Interfacial tension model

There are two types of interfacial models: CSF model developed by Nomura et al.\textsuperscript{18}), which considers volume force, and the potential model proposed by Kondo et al.\textsuperscript{19}), which formalizes intermolecular force. However, these models are not suitable for liquid-liquid two-phase flow because they consider only the force in single fluid. This paper therefore adopts the interfacial tension model proposed by Ishii et al.\textsuperscript{5}), which considers the influence of the other phase in two-phase flow. The model is based on the potential model developed by Kondo et al.\textsuperscript{19}) and is defined as follows.

\begin{equation}
F_i = C \sum_{j \neq i} f_{ij} \frac{(r_j - r_i)}{r_{ij}}
\end{equation}

where $F_i$ is the potential force of particle $i$, $C$ is the potential coefficient, and $f_{ij}$ is a function that calculates the potential force between particles $i$ and $j$. The potential force function $f_{ij}$ is defined in the following.

\begin{equation}
f_{ij}(r_{ij}) = \begin{cases} (r_{ij} - l_0) & (r_{ij} \leq r_e) \\ 0 & (r_{ij} > r_e) \end{cases}
\end{equation}

where $l_0$ is the initial distance between particles. The pressure in the normal direction is calculated as follows.

\begin{equation}
p_{i}^n = \frac{F_i \cdot n_i}{A_i}
\end{equation}

where $P_i^n$ is the pressure in the normal direction of particle $i$, and $n_i$ is the normal unit vector of particle $i$, and $A_i$ is the small area element of particle $i$. Here, $n_i$ is calculated in the following.

\begin{equation}
n_i = \begin{cases} F_i / |F_i| & (|F_i| / |F_i|_{flat} \geq \gamma) \\ 0 & (|F_i| / |F_i|_{flat} < \gamma) \end{cases}
\end{equation}

where $|F_i|_{flat}$ is the magnitude of the potential force at zero interfacial curvature and $\gamma$ is the interfacial decision coefficient, which was set as 0.2 by Ishii et al.\textsuperscript{5}). However, the normal vector is calculated only for the particles near the interface, and it becomes a zero vector by using Eq. (14) for the internal particles. In addition, the small area element $A_i$ for particle $i$ is calculated as follows.

\begin{equation}
A_i = A_0 \frac{1}{N} \sum_{j \neq i} \sin \phi_{ij}
\end{equation}

where $A_0$ is the interfacial area occupied by one particle, $N$ is the particle number in the radius of influence, and $\phi_{ij}$ is the angle between the normal vectors of particles $i$ and $j$.

The volume force $F_{inter}$ of interface particle $i$ is obtained as the pressure gradient $\nabla P_i^n$ in the following.

\begin{equation}
F_{inter} = \nabla p_i^n = \frac{d(P_{liquid} - P_{gas})}{\hat{h}} n_i
\end{equation}

where $F_{inter}$ is the volume force of the particle $i$, $P_{liquid}$ is the pressure of the water phase, $P_{gas}$ is the pressure of the gas phase, and $\hat{h}$ is the interfacial thickness, which equals the initial particle distance.

The interfacial tension model developed by Ishii et al.\textsuperscript{5}), is formulated assuming gas-liquid two-phase flow instead of liquid-liquid flow. Therefore, they considered that the pressures of liquid-gas phases were balanced at the
interface with zero curvature and extrapolated the pressure of the gas phase as that of the liquid phase with zero curvature without calculation for the gas phase. The equation to calculate the volume force $F_{i\text{inter}}$ is defined as follows, where $P_{gas}$ in Eq. (16) is replaced with $P|_{flat}$ in Eq. (17). Furthermore, $P|_{flat}$ is the same as $P^i$ in Eq. (13) when the curvature is zero. It is possible to extrapolate the pressure with Eq. (17) without calculating $P_{gas}$ by transforming Eqs. (13) - (16).

$$F_{i\text{inter}} = \frac{d(P_{\text{liquid}} - P|_{\text{flat}})}{h} n_i$$

where $P|_{\text{flat}}$ is the liquid phase pressure with zero curvature. Figure 1 illustrates the idea of Eq. (17).

In Fig. 1, the black and white particles are liquid and gas particles, respectively, and the dotted line is the interface between liquid and gas. In this study, we consider liquid-liquid two-phase flow instead of liquid-gas phase flow. Then, it is not necessary to extrapolate gas phase pressure because the liquid phase corresponds to the gas phase. We propose the following formula by which to calculate the volume force for liquid-liquid two-phase flow of viscous fluid and water. Here, viscous fluid is a water-immiscible liquid and excludes water.

$$F_{i\text{inter}}^v = \frac{d(P_v - \hat{P}_w)}{h} n_i$$

where $P_v$ is the pressure of viscous fluid and $\hat{P}_w$ is the average pressure of water in the radius of influence. $\hat{P}_w$ is defined in the following.

$$\hat{P}_w = \frac{1}{N'} \sum_{j \in i} P_w \omega'(r_{ij})$$

where $r_{ave}^w$ is the radius of influence used for the calculation of the average pressure of viscous fluid, $N'$ is the number of particles in $r_{ave}^w$, and $\omega'$ is the weight function. $r_{ave}^w$ equals the interfacial thickness $h$ and is set as the initial particle distance $l_0$. Figure 2 shows the idea of Eq. (18).

In Fig. 2, the black and white particles are viscous and water particles, respectively. The black circle is the radius of influence used for the calculation of $\hat{P}_w$. If there is no particle of the other phase within $r_{ave}^w$, the volume force becomes huge because $\hat{P}_w$ is zero and the gradient of Eq. (18) becomes large. In this case, $\hat{P}_w$ is replaced with $P|_{\text{flat}}$ in Eq. (18) to prevent the volume force from being huge.

### 3.5 Implicit method for viscous term

MPS calculation procedure developed by Koshizuka et al. is a semi-implicit method, in which the process, viscosity, gravity acceleration, and interfacial tension terms are solved using the explicit method, and then the pressure term is solved using the implicit method. In this procedure, the time step $\Delta t$ should be very small because kinematic viscosity $\nu$ is very high in von Neumann condition, which is defined as follows.

$$\Delta t \leq \frac{l_0}{2\nu}$$

In this study, since the viscosity of viscous fluid injected from a circular tube is about 600 times that of water, the time step should be about 1/600 times, and the analysis takes a huge amount of time. We therefore apply the implicit method proposed by Fukuzawa et al. to calculate the viscous term, allowing us to set the time step as a large value. The implicit formula for the viscous term is defined in the following.

$${u^i_k - u^i} \over \Delta t = \nu \nabla^2 u^i + g + {1 \over \rho} F_{i\text{inter}}$$

where $u^i$ is the velocity vector of particle $i$ at the time step $k$. In our procedure, the gravity acceleration and the interfacial tension terms are solved by the explicit method, and then the viscous and the pressure terms are solved by the implicit method. In addition, we adopt BI-CGSTAB as the solver for the implicit calculation of Eqs. (9) and (22).

### 4. Results

#### 4.1 Simulation condition

Table 1 shows the specifications of the workstation.
used for the simulation.

Table 2 shows the conditions for both of the physical experiment and the simulation.

The two conditions with an asterisk (*) in Table 2 are applied only to the simulation.

Figure 3 shows the environments of the physical experiment for the quantitative assessment and the simulation model.

Note that the outflow boundary is set at 0.2 [mm] below the upper boundary of the water tank in Fig. 3 (b) and that the nonslip condition is applied to the walls of the water tank and the circular tube.

4.2 Simulation and physical experiment results

Figures 4 and 5 show the potential model developed by Kondo et al.19 and the proposed model in this paper, as well as the physical experiment results. For the timing of viscous fluid injection in the physical experiment, the first time taken from droplet formation to the dripping is different from the time of the subsequent droplets since the inflow velocity is not stable at the beginning. We therefore excluded the first droplet in the physical experiment from the comparison. In addition, residual droplets are generated after the droplet is detached from the tube. For the comparison considering the residual droplets, we calculated the volume of the residual droplets immediately after the first droplet fell from the tube in the physical experiment. As a result, we found that the volume of the residual droplet corresponds to the volume injected during 1.03 seconds in a stable state. Then, Figs. 4 and 5 show the results of the

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Unit</th>
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<tbody>
<tr>
<td>Density</td>
<td></td>
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</tr>
<tr>
<td>water</td>
<td>$1.00 \times 10^3$</td>
<td>kg/m$^3$</td>
</tr>
<tr>
<td>viscous fluid</td>
<td>$1.182 \times 10^3$</td>
<td>kg/m$^3$</td>
</tr>
<tr>
<td>Kinematic viscosity</td>
<td></td>
<td></td>
</tr>
<tr>
<td>water</td>
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<td>m$^2$/s</td>
</tr>
<tr>
<td>viscous fluid</td>
<td>$628.43 \times 10^{-6}$</td>
<td>m$^2$/s</td>
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<td>Gravity acceleration</td>
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<td>Interfacial tension coefficient</td>
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</tr>
<tr>
<td>Injection velocity</td>
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<td>m/s</td>
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<tr>
<td>Volume flow rate</td>
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<td>m$^3$/s</td>
</tr>
<tr>
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</tr>
<tr>
<td>Time step</td>
<td>$1.00 \times 10^{-4}$</td>
<td>s</td>
</tr>
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Table 2 Physical experiment and simulation conditions

Fig. 3 Environment of the physical experiment and the simulation model [mm]

Fig. 4 Simulation and physical experiment results between the time
potential model, the proposed model, and the physical experiment from 1.03 s. Here, the water particles in the water tank are not visualized in order to improve the visibility of the injected viscous fluid, and black particles represent the viscous fluid. Three images at 1.03 s in Fig. 4 show the initial states for (a), (b), and (c). The droplets grow over time. Details of the comparison of the proposed model with the potential model and with the physical experiment are described in Sections 4.3 and 4.4, respectively.

Figure 5 is a continuation from the time at 17.00 s in Fig. 4.

4.3 Comparison of the simulation methods

Figure 6 compares the potential model developed by Kondo et al.\textsuperscript{19)} and the model proposed in this paper. Note that both simulations were performed under the same conditions. From Fig. 6 (a) and (b), the droplets in the potential and proposed models differ in shape at 6.0 s. In Fig. 6 (a), the droplet does not touch the tip of the tube, whereas in Fig. 6 (b) it does touch the tip. At 10.0 s, the droplet in Fig. 6 (a) grows without changing shape. On the other hand, the middle of the droplet in Fig. 6 (b) has a neck. At 14.0 s, the droplet in Fig. 6 (a) is not detached from the tip of the tube, whereas it is detached in Fig. 6 (b). The detached time in the proposed model is 12.20 s and 15.30 s in the potential model. The difference seems attributable to the influence of the other phase in two-phase flow. The potential model, which does not take into account the influence of force from the other phase, takes time to change the shape of the droplet, while the proposed model, which considers the influence of the other phase, can change the shape of the droplet by considering the force from the other phase. This is why the detached times differ between the two models.

Figure 7 shows magnified views of Fig. 6. The tip of the tube is emphasized by a black line for clarification. In Fig. 7, it can be seen that the droplet in the potential model does not contact with the tip of the tube at 1.03 s, whereas the droplet in the proposed model contacts the tip at 1.03 s and 8.00 s.
4.4 Comparison of physical experiment and simulation

Figure 8 compares the physical experiment and the simulation by the proposed method.

Then, Fig. 8 shows the states after the first droplet is detached from the tube. In addition, some viscous fluid remains in the tube after the first droplet has been detached, and the volume has the same amount as the droplet after 1.03 [s] from the beginning of the simulation. Therefore, Fig. 8 shows the states after 1.03 [s]. In Fig. 8 (a) and (b), we can see that the droplets formed by the simulation grow vertically at 4.00 [s] and 8.00 [s], and a slight neck appears at 11.00 [s].

Figure 9 shows the magnified views of Fig. 8. The tip of the tube is emphasized by a black line for clarification. In the physical experiment of Fig. 9 (a), the viscous fluid is slightly wetted at the tip of the tube, whereas it is not wetted in the simulation of Fig. 9 (b).

Figure 10 shows the second droplets in the physical experiment and the simulation. Note that the comparison starts at 1.03 [s] in Fig. 4 where one droplet is already at the bottom of the water tank in the physical experiment because of unstable inflow for the first droplet in the physical experiment.

In Fig. 10 (a), the viscous fluid that remains at the tip of the tube is more elongated vertically, whereas it is not elongated vertically in Fig. 10 (b). This elongation is due to the polymer fluid structure; the polymer behavior of viscous fluid is not considered in the simulation.

In addition, Table 3 shows the detached times of the physical experiments and the simulations by the
proposed and potential models. Note that the physical experiment has 6 droplets, whereas the simulations in the proposed and potential models have 3.

In Table 3, the detached time difference of the proposed model was smaller than that of the potential model.

Table 4 shows the drop cycle times of the physical experiments and the simulations by the proposed and the potential models.

As Table 4 shows, the average drop cycle in the experiment was 11.35 [s]. On the other hand, in the simulation the drop cycles for the proposed and potential models were 11.57 [s] and 13.16 [s], respectively. Thus, the drop cycle of the proposed model is much closer to that of the physical experiment than that of the potential model. Therefore, it is considered that the influence of the other phase is important.

On the other hand, Figs. 11 and 12 show the averages shape changes of the droplets for the physical experiment and the simulation by the proposed model in width and length, respectively. The left axis shows the physical length [mm], while the right axis shows the ratio, which is calculated by dividing the droplet length in the simulation by that in the physical experiment. From Fig. 11, we can see that the width changes of the droplets in the physical experiment and the simulation by the proposed model are very close all the time. In Fig. 13, however, the length changes are different because the droplet in the simulation is elongated vertically.

5. Conclusions

In this study, we have proposed a new droplet formation method for liquid-liquid two-phase flow, which is a modification of the model developed by Ishii et al. We have also examined the validity of the potential model developed by Kondo et al. and that of our proposed method. The comparison revealed that the droplets did not contact with the tip of the tube in the potential model developed by Kondo et al., whereas they did contact the tip in the proposed model. In addition, the comparison of the physical experiment and the proposed method showed that both drop cycles are very close and that the droplet width changes are almost the same between the physical experiment and the simulation, whereas the droplet length changes differ because the behavior of a polymer fluid structure was not considered in the simulation. The polymer fluid is a pseudoplastic, and the viscosity coefficient decreases as the velocity gradient increases. Thus, the method that has a constant viscosity coefficient is not enough for the simulation of polymer fluid. In addition, the study of particle method that can deal with pseudoplastic fluid has been hardly researched, so a useful method for liquid-liquid two phase flow simulation has not been established. In addition, in a simulation where a viscous

<table>
<thead>
<tr>
<th>Table 3 Droplet detached times</th>
<th>[s]</th>
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<tr>
<td>Physical experiment (1) Simulations</td>
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<tr>
<td>Proposed</td>
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<tr>
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<table>
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<th>Table 4 Drop cycle times</th>
<th>[s]</th>
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<td>Drop cycle number</td>
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<tr>
<td>1</td>
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<tr>
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<td>5</td>
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<td>Average</td>
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fluid is injected into another type of fluid, the interfacial tension and the viscosity are very important. We have solved the viscous term of the Navier-Stokes equation implicitly in order to handle highly viscous fluid. However, it is very difficult to control the interfacial tension because the droplet formation process changes depending on the interfacial tension model. Therefore, the proposed model can be applied to a droplet simulation in which viscosity changes, whereas it is not suitable for a simulation where the interfacial tension changes.

In the future, therefore, we have to establish a method that can deal with polymer fluid and perform a simulation in which the droplet shape is closer to that in the physical experiment.

References